S1. Derivation of the concentration fields from the simulations using synthetic meteorological data

Although the wind direction was randomised in the meteorological data used in the short-range dispersion modelling, the resulting concentration fields were not symmetrical due to the influence of short-term events such as very stable periods. In order to obtain generic rotationally symmetric concentration fields that can be used in the sub-grid model, the asymmetry was removed by fitting polynomial regression curves, of the form $ax^3 + bx^2 + cx + d$, to the modelled concentrations (natural log of concentrations vs. natural log of distance (m) from source centre), as shown in Figure A2.1. For each source height, the mean value of the predictions from all regression curves was used to calculate the concentration fields used in the sub-grid model for synthetic meteorology (Table A2.1).

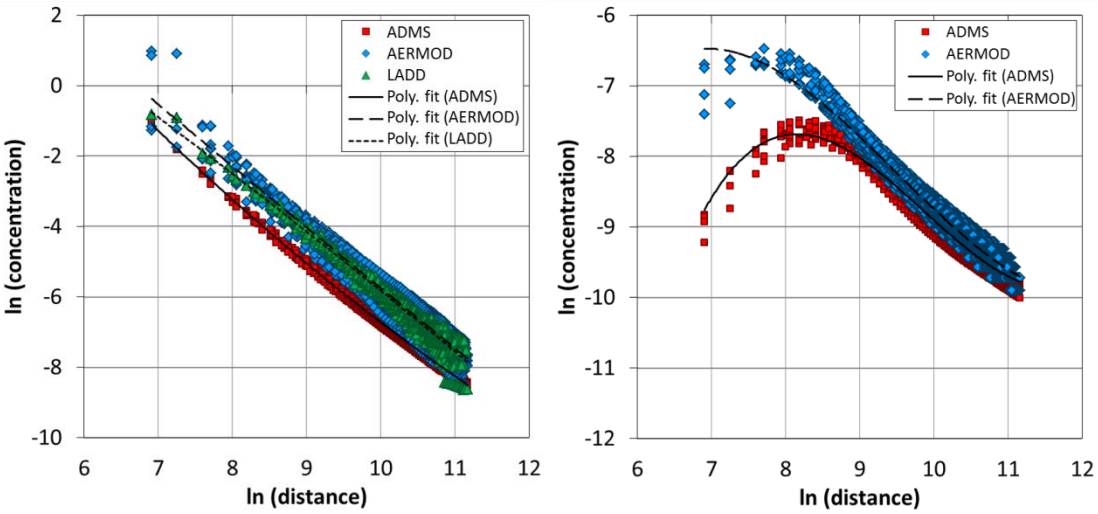


Figure S1.1: Natural log of modelled concentration plotted against the natural log of distance from source centre for a ground level source (left) and a 400 m high stack source (right).

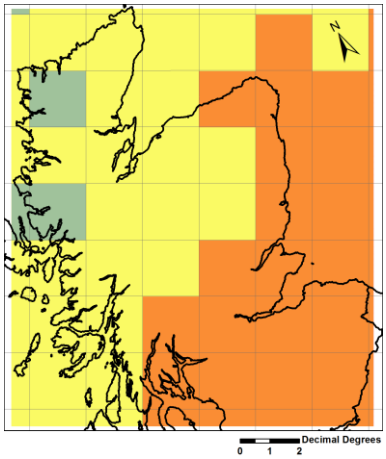
Table S1.1: Regression coefficients of the polynomial fits (of the form: $ax^3 + bx^2 + cx + d$) to the natural log of modelled concentration vs. the natural log of distance (m) from the source centre for all source heights and models.

Emission height: 0 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	--	-0.0356	-0.964	7.46	1.69
AERMOD	--	0.0747	-3.08	16.6	0.864
LADD	--	-0.0189	-1.32	9.28	1.32
Mean model	--	--	-1.67	10.7	1.34
Emission height: 25 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	-0.0192	0.573	-7.03	25.5	-0.895
AERMOD	-0.0261	0.602	-5.56	15.9	-1.88
Mean model	-0.0295	0.754	-7.58	23.8	-1.27
Emission height: 50 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	0.0109	-0.294	1.28	-1.40	-2.35
AERMOD	-0.0241	0.692	-7.56	24.1	-3.02
Mean model	-0.0105	0.318	-4.31	15.1	-2.63
Emission height: 100 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	0.00850	-0.259	1.49	-4.84	-4.45
AERMOD	-0.0213	0.572	-6.00	16.8	-4.02
Mean model	-0.00610	0.149	-2.22	5.96	-4.22
Emission height: 200 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	0.0283	-0.813	6.85	-23.5	-6.88
AERMOD	-0.0290	0.885	-9.80	30.3	-5.39
Mean model	-0.00150	0.0705	-1.82	4.58	-5.88
Emission height: 400 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	0.108	-3.20	30.6	-103	-12.7
AERMOD	0.0532	-1.52	13.4	-44.0	-7.21
Mean model	0.0666	-1.93	17.7	-59.3	-7.90
Emission height: 800 m					
Model	**a**	**b**	**c**	**d**	**Value for emission square**
ADMS	0.189	-6.00	62.7	-225	-51.0
AERMOD	0.109	-3.30	32.3	-111	-9.90
Mean model	0.0900	-2.81	28.2	-101	-10.6

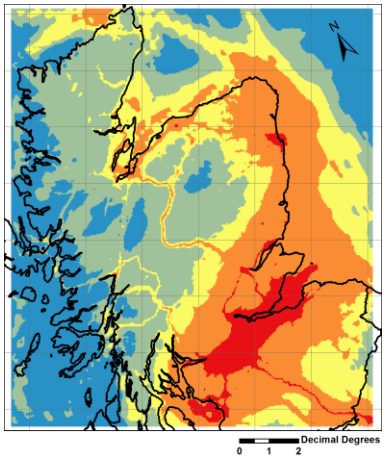
S2. Converting “sub-grid distributions” to “sub-grid concentrations”

Example: Annual mean NO_2 concentrations for the central Scotland domain

Starting datasets:



EMEP annual mean NO_2 concentration (50 × 50 km^2)

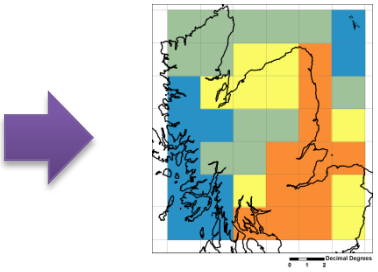


Sub-grid annual mean NO_2 distribution (1 × 1 km^2)

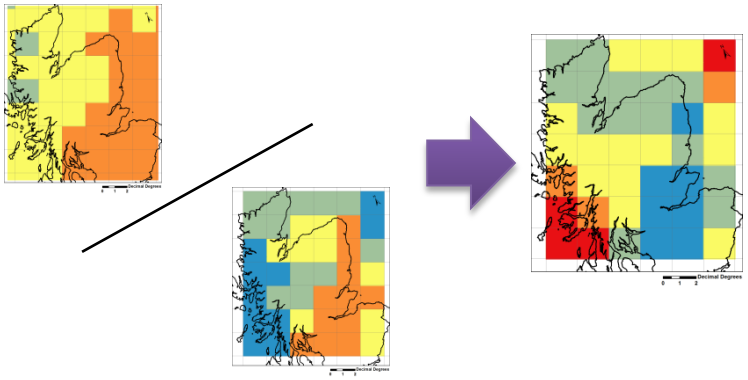
Process flowchart

START

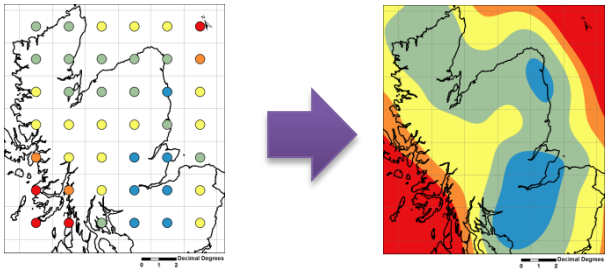
Calculate 50 × 50 km^2 mean values of the sub-grid distribution



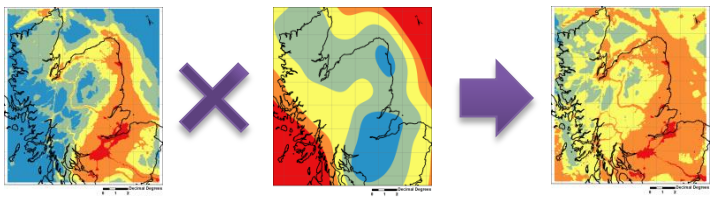
Divide EMEP concentrations by the mean values of the sub-grid distribution



Interpolate these ratios at the 50 km grid cell centres across the domain using cubic splines



Multiply these interpolated values by the sub-grid distribution to produce a new sub-grid distribution

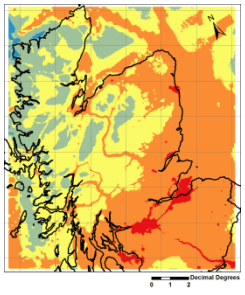


10 Iterations completed?

No

Yes

END



After 10 iterations

S3. Model evaluation

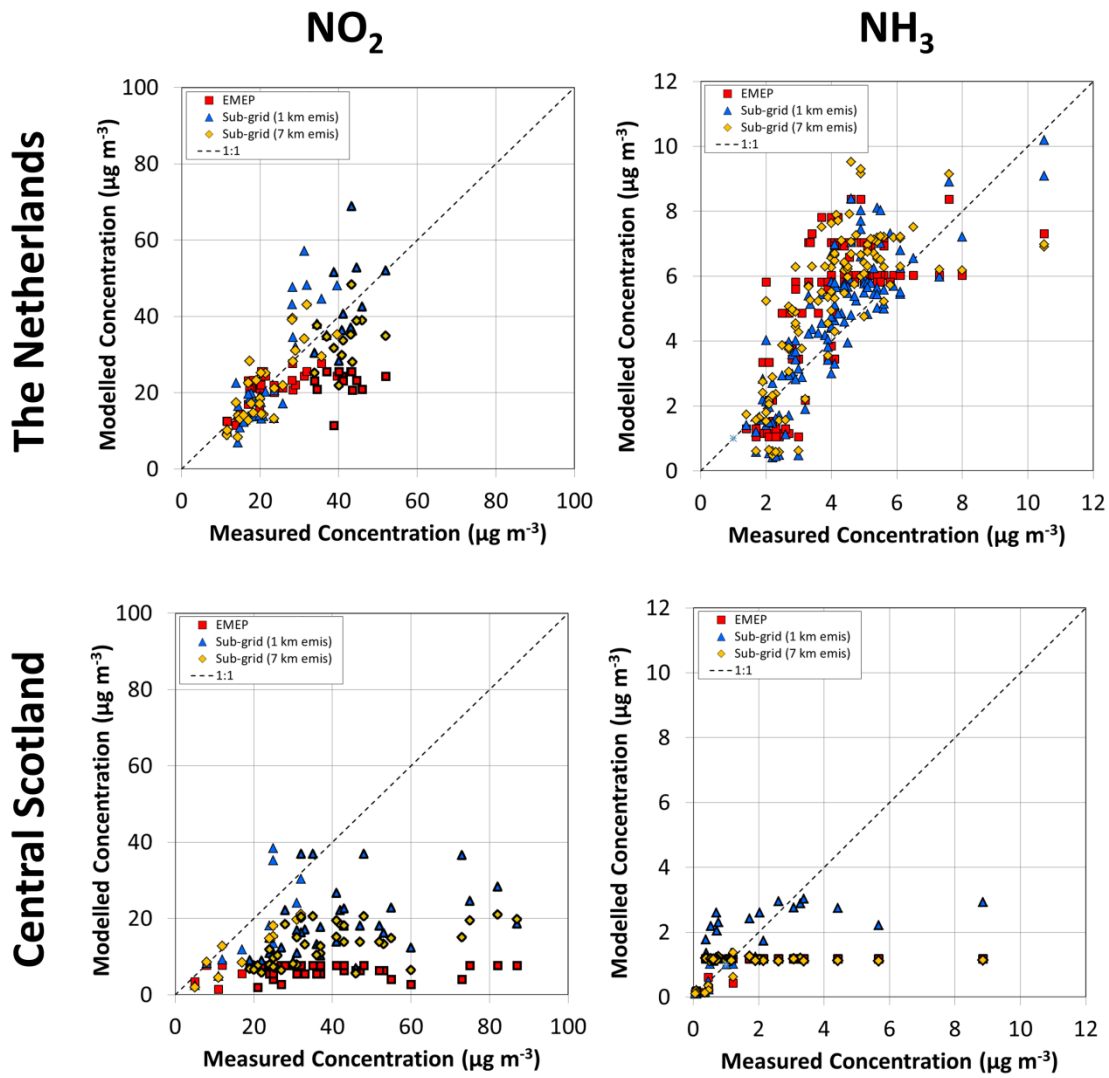


Figure S3.1: Modelled concentrations plotted against measured values for all sites for NO_2 (left column) and NH_3 (right column) for the Dutch (top row) and Scottish (bottom row) domains. NO_2 traffic stations and the NH_3 local network are indicated by bold symbol outlines.